

CARBON DIFFUSION

Project Staff

General Atomics
3550 General Atomics Court
San Diego, CA 92121-1194

November 1996

Final Report

Distribution authorized to DoD and U.S. DoD contractors only; Critical Technology; November 1996. Other requests for this document shall be referred to Phillips Laboratory/VTVP, 3550 Aberdeen Ave. SE, Kirtland AFB, NM 87117-5776.

WARNING - This document contains technical data whose export is restricted by the Arms Export Control Act (Title 22, U.S.C., Sec 2751 *et seq.*) or The Export Administration Act of 1979, as amended (Title 50, U.S.C., App. 2401, *et seq.*). Violations of these export laws are subject to severe criminal penalties. Disseminate IAW the provisions of DoD Directive 5230.25 and AFI 61-204.

DESTRUCTION NOTICE - For classified documents, follow the procedures in DoD 5200.22-M, Industrial Security Manual, Section II-19 or DoD 5200.1-R, Information Security Program Regulation, Chapter IX. For unclassified, limited documents, destroy by any method that will prevent disclosure of contents or reconstruction of the document.

19970210 155



PHILLIPS LABORATORY
Space Technology Directorate
AIR FORCE MATERIEL COMMAND
KIRTLAND AIR FORCE BASE, NM 87117-5776

UNCLASSIFIED



AD NUMBER

AD-B219 965

NEW LIMITATION CHANGE

TO

DISTRIBUTION STATEMENT A -

Approved for public release; Distribution unlimited.

Limitation Code: 1

FROM

DISTRIBUTION STATEMENT -

Limitation Code:

AUTHORITY

**Janet E. Mosher; Phillips Lab/CA, Kirtland AFB,
N.M.**

THIS PAGE IS UNCLASSIFIED

PL-TR-96-1146

Using Government drawings, specifications, or other data included in this document for any purpose other than Government procurement does not in any way obligate the U.S. Government. The fact that the Government formulated or supplied the drawings, specifications, or other data, does not license the holder or any other person or corporation; or convey any rights or permission to manufacture, use, or sell any patented invention that may relate to them.

If you change your address, wish to be removed from this mailing list, or your organization no longer employs the addressee, please notify PL/VTV, 3550 Aberdeen Ave SE, Kirtland AFB, NM 87117-5776.

Do not return copies of this report unless contractual obligations or notice on a specific document requires its return.

This report has been approved for publication.



**DR. MICHAEL SCHULLER
Project Manager**

FOR THE COMMANDER



**L. KEVIN SLIMAK, GM-15
Chief, Space Vehicle Technologies
Division**



**CHRISTINE M. ANDERSON
Director, Space Technology
Directorate**

The following notice applies to any unclassified (including originally classified and now declassified) technical reports released to "qualified U.S. contractors" under the provisions of DoD Directive 5230.25, Withholding of Unclassified Technical Data From Public Disclosure.

NOTICE TO ACCOMPANY THE DISSEMINATION OF EXPORT-CONTROLLED TECHNICAL DATA

1. Export of information contained herein, which includes, in some circumstances, release to foreign nationals within the United States, without first obtaining approval or license from the Department of State for items controlled by the International Traffic in Arms Regulations (ITAR), or the Department of Commerce for items controlled by the Export Administration Regulations (EAR), may constitute a violation of law.
2. Under 22 U.S.C. 2778 the penalty for unlawful export of items or information controlled under the ITAR is up to two years imprisonment, or a fine of \$100,000, or both. Under 50 U.S.C., Appendix 2410, the penalty for unlawful export of items or information controlled under the EAR is a fine of up to \$1,000,000, or five times the value of the exports, whichever is greater; or for an individual, imprisonment of up to 10 years, or a fine of up to \$250,000, or both.
3. In accordance with your certification that establishes you as a "qualified U.S. Contractor", unauthorized dissemination of this information is prohibited and may result in disqualification as a qualified U.S. contractor, and may be considered in determining your eligibility for future contracts with the Department of Defense.
4. The U.S. Government assumes no liability for direct patent infringement, or contributory patent infringement or misuse of technical data.
5. The U.S. Government does not warrant the adequacy, accuracy, currency, or completeness of the technical data.
6. The U.S. Government assumes no liability for loss, damage, or injury resulting from manufacture or use for any purpose of any product, article, system, or material involving reliance upon any or all technical data furnished in response to the request for technical data.
7. If the technical data furnished by the Government will be used for commercial manufacturing or other profit potential, a license for such use may be necessary. Any payments made in support of the request for data do not include or involve any license rights.
8. A copy of this notice shall be provided with any partial or complete reproduction of these data that are provided to qualified U.S. contractors.

D E S T R U C T I O N N O T I C E

For classified documents, follow the procedures in DoD 5200.22-M, Industrial Security Manual, Section II-19 or DoD 5200.1-R, Information Security Program Regulation, Chapter IX. For unclassified, limited documents, destroy by any method that will prevent disclosure of contents or reconstruction of the document.

DRAFT SF 298

1. Report Date (dd-mm-yy) November 1996	2. Report Type Final	3. Dates covered (from... to) 01/92 to 08/96			
4. Title & subtitle Carbon Diffusion		5a. Contract or Grant # F29601-92-C-0057			
		5b. Program Element # 63401F			
6. Author(s) Project Staff		5c. Project # 3977			
		5d. Task # TE			
		5e. Work Unit # AH			
7. Performing Organization Name & Address General Atomics 3550 General Atomics Court San Diego, CA 92121-1194		8. Performing Organization Report #			
9. Sponsoring/Monitoring Agency Name & Address Phillips Laboratory 3550 Aberdeen Ave SE Kirtland AFB, NM 87117-5776		10. Monitor Acronym			
		11. Monitor Report # PL-TR-96-1146			
12. Distribution/Availability Statement Distribution authorized to DoD and U.S. DoD contractors only; Critical Technology; November 1996. Other requests for this document shall be referred to Phillips Laboratory/VTVP, 3550 Aberdeen Ave SE, Kirtland AFB, NM 87117-5776. <i>Kirtland AFB, NM 87117-5776</i>					
13. Supplementary Notes					
14. Abstract In the STAR-C out-of-core space nuclear power system, heat is radiated from a graphite core onto the emitter/hot shoe of thermionic converters. Carbon evaporates from the surface of the core and condenses on the emitter/hot shoe. There is concern that this carbon may diffuse through the emitter/hot shoe, reaching the emitter surface. This may alter the work function of the emitter and degrade electrical output. In this work, a carbon source was placed directly facing an uncoated, chemical vapor deposition (CVD) polycrystalline tungsten specimen. The bare work function of the back side of the tungsten specimen (not facing the carbon source) was measured before testing. Both the carbon source and tungsten specimen were heated and a fluence of carbon was allowed to deposit onto the adjacent tungsten surface. Under accelerated conditions, a fluence corresponding to 255 days of operation, at the STAR-C system operating conditions, was deposited onto the tungsten specimen. Work function measurements of the back side of the tungsten revealed no detectable change in the bare work function.					
15. Subject Terms Thermionics; Carbon Diffusion; Energy Conversion					
Security Classification of		19. Limitation of Abstract	20. # of Pages	21. Responsible Person (Name and Telephone #)	
16. Report Unclassified	17. Abstract Unclassified	18. This Page Unclassified	Limited	24	Dr. Michael Schuller (505) 846-4945

CONTENTS

<u>Section</u>	<u>Page</u>
1.0 OBJECTIVE AND TECHNICAL APPROACH	1
2.0 EXPERIMENTAL SETUP	3
3.0 INITIAL SYSTEM CHECKOUT	8
4.0 TEST RESULTS	12
5.0 DISCUSSION AND CONCLUSIONS	13

FIGURES

<u>Figure</u>	<u>Page</u>
1. Experimental Setup.	3
2. Front views of tungsten button/heat choke subassembly (left) and guarded electrode subassembly.	5
3. Rear views of tungsten button/heat choke subassembly (left) and guarded electrode subassembly (right).	6
4. Pre-assembled geometry of guarded electrode subassembly mating with the tungsten button/heat choke subassembly.	7
5. Carbon source subassembly.	9
6. Carbon evaporation test assembly.	9
7. Test schematic.	10
8A. Revised experimental setup.	11
8B. Setup during work function measurement.	11
9. Carbon concentration vs time: experiment vs STAR-C conditions.	14

TABLES

<u>Table</u>	<u>Page</u>
1. Component temperatures: experiment vs STAR-C conditions.	13

1.0 OBJECTIVE AND TECHNICAL APPROACH

The purpose of this work was to produce and test materials and coatings which would limit the ingress of carbon into the tungsten hot shoe/emitters of thermionic converters in ex-core reactor designs.

In the STAR-C space nuclear power system, for example, heat radiates from a graphite core to the tungsten hot shoes of the surrounding converters. In this configuration, carbon evaporates from the surface of the core and condenses on the tungsten hot shoes and may diffuse through the tungsten to the electron-emitting surface in the interelectrode gap of the converter. The presence of carbon on this emitter surface may adversely affect the work function, degrading thermionic performance. This has been identified as a key feasibility issue of the graphite core design.

The work was planned in sequential phases. The first step involved heating a carbon source to evaporate carbon onto the surface of an uncoated, polycrystalline tungsten sample. The second step was the measurement of the work function on the opposite side of the tungsten sample to track changes in work function. The plan was to achieve an equivalent of a 10 yr exposure (at STAR-C operating conditions). After work function measurements were made, the tungsten sample would be sectioned and the carbon concentration would be measured through the thickness. The results would become a baseline against which subsequent results could be compared.

In the next step, the above procedure was to be repeated using a single crystal tungsten sample. Since most of the carbon diffusion was believed to occur along the grain boundaries within the tungsten matrix, it was believed that the absence of grain boundaries in single crystal material would reduce the carbon diffusion

rate. This would reduce the carbon concentration on the emitting side of the hot shoe-emitter.

Subsequent steps would have involved applying carbon diffusion-resistant coatings, such as NbC, HfC, to a tungsten sample and repeating the experiment.

The results of this work would have provided a comparison of the carbon concentration, and its effect on work function, for three conditions: uncoated, polycrystalline tungsten; uncoated single crystal tungsten; and coated tungsten systems.

2.0 EXPERIMENTAL SETUP

A schematic of the conceptual setup is shown in figure 1. A carbon source was heated to as high as 2200 K by an electron bombardment filament. The heated carbon source was the source of carbon flux to a tungsten emitter sample. Radiant heating from the carbon source also heated the tungsten sample up to 1950 K. Located on the opposite side of the tungsten button was a collector for measuring the electron emission from the button, from which the work function could be determined. This collector was surrounded by, and isolated from, a guard ring. This guard ring establishes the exact area over which emission current is collected. An existing GA sheath insulator assembly was used for the collector/guard ring.

CARBON DIFFUSION TEST EXPERIMENTAL SETUP

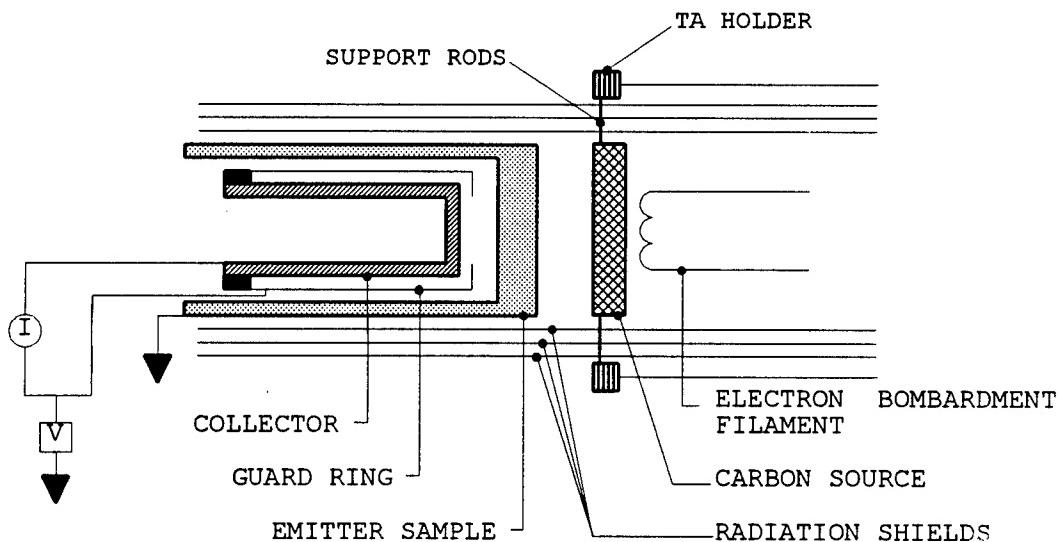


Figure 1. Experimental setup.

An existing vacuum station was located and dedicated exclusively to this experiment. The station was pumped by a turbomolecular pump. In the event of an after-hours power failure, the turbomolecular pump would slowly rise up to atmospheric pressure. If this occurred, the test pieces could become contaminated within the vacuum station. To prevent this from happening, an air-actuated gate valve was installed between the turbomolecular pump and the vacuum station as a safety precaution. In the event of a power failure, the valve would fail shut, isolating the test chamber from the pump thereby protecting the experiment from inadvertent exposure to the atmosphere. The vacuum station was bakeable and capable of attaining a base vacuum of 6×10^{-9} torr.

A thermal analysis of the experimental setup was performed. Dimensions of the emitter stem were sized to limit conduction heat loss to allowable levels. Power requirements for the bombardment filament were determined and a high voltage power supply was obtained.

Several [110] oriented CVD tungsten emitter buttons were on hand at GA and were used as the polycrystalline tungsten samples. A fabrication technique was developed to CVD tungsten stems onto these button samples. The tungsten button was mounted onto a mandrel onto which tungsten was vapor deposited. After the CVD operation, the emitter stem OD was ground to provide the specified stem thickness and the sacrificial mandrel, onto which the stem was CVD'd, was etched away. A hohlraum, for use with an optical pyrometer, was then EDM'd into the emitter assembly. The emitter heat choke/Ta/SS transition pieces initially proved difficult to braze together. A susceptor to enhance RF coupling to the SS part was built and the braze was successful. The collector/guard ring/trilayer assembly was electron beam welded together.

The test subassembly, consisting of the carbon source, electron bombardment

filament and heat shields was installed into the vacuum system for verification of operation.

Figures 2 and 3 show front and rear views, respectively, of the emitter button and CVD'd heat choke subassembly (left) and the guarded electrode/collector subassembly (right). As shown in Figure 4, the guarded electrode subassembly fits within the emitter button/heat choke subassembly, directly opposite the back side of the polycrystalline tungsten emitter button. Using this setup, work function measurements could be made.

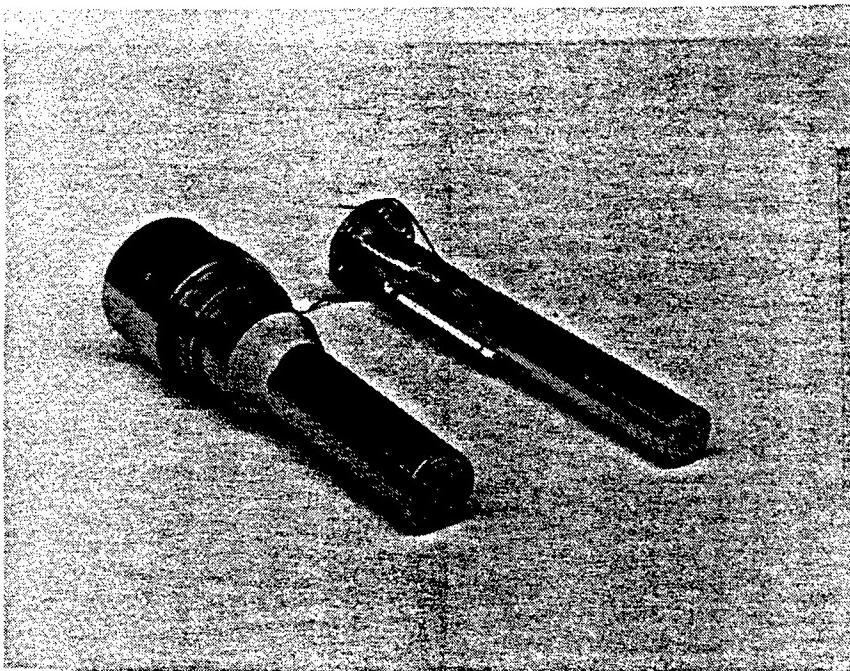


Figure 2. Front views of tungsten button/heat choke subassembly (left) and guarded electrode subassembly.

A domestic source of single crystal tungsten of the size required for this effort could not be found. Russian sources did exist which could custom fabricate single crystal tungsten specimens to our specifications. After approval was gained from DoD, a purchase order for a 2-cm diameter single crystal tungsten disk was procured from the Scientific Institute LUTCH, in Russia.

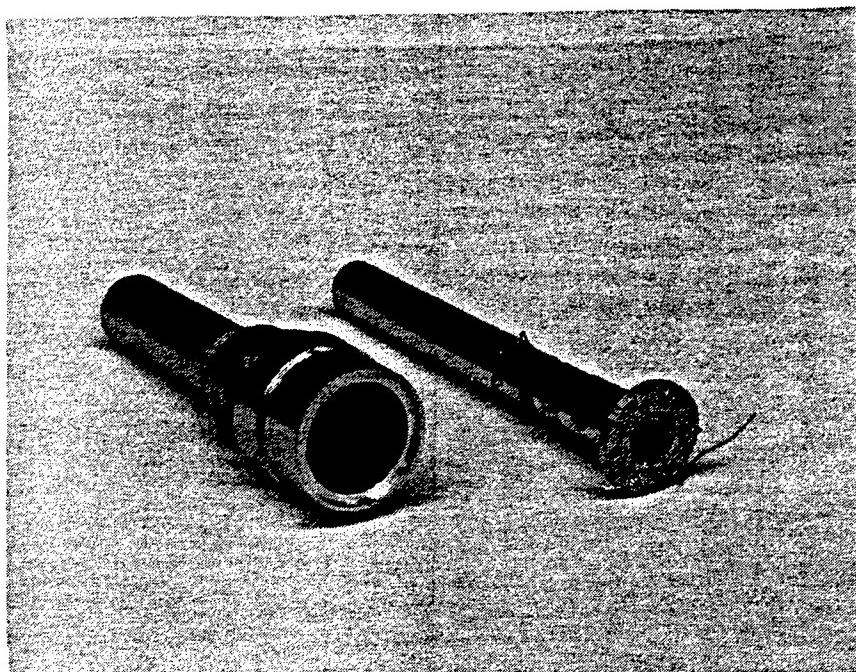


Figure 3. Rear views of tungsten button/heat choke subassembly (left) and guarded electrode subassembly (right).

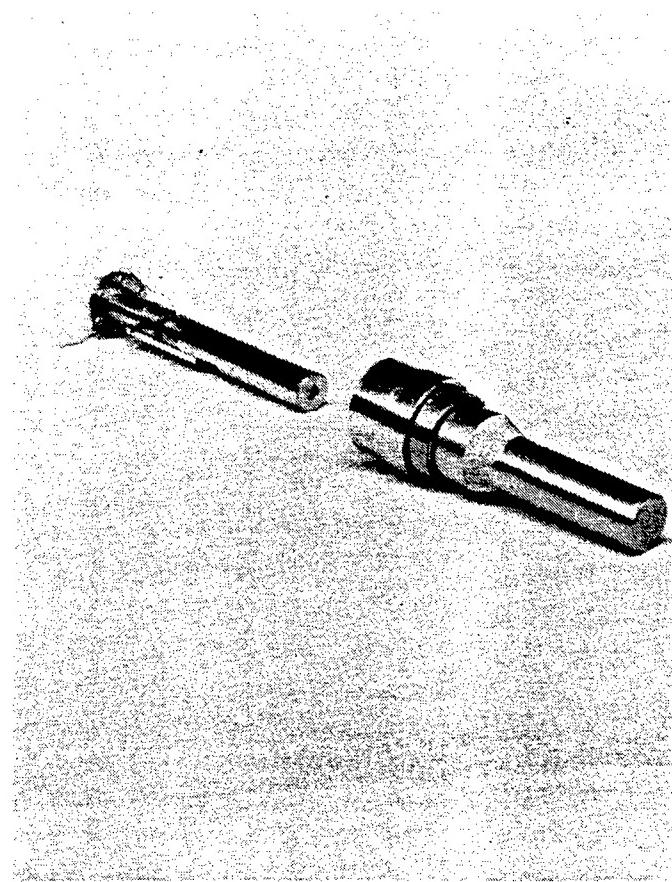


Figure 4. Pre-assembled geometry of guarded electrode subassembly mating with the tungsten button/heat choke subassembly.

3.0 INITIAL SYSTEM CHECKOUT

Before initial checkout started, the turbomolecular pump used in the vacuum system broke down and was replaced by an ion pump. The carbon source subassembly was placed in the vacuum system and heated by electron bombardment heating. During this checkout heating, the rods used to center the carbon source within the Ta holder and the rods used to suspend the Ta holder within the test chamber were found to conduct too much heat away from the carbon source. The diameter of both sets of rods was turned down to 1/16" dia to reduce their conduction losses.

Two electron bombardment power supplies broke down during heat up testing of the carbon source. A third electron bombardment power supply was installed and the system checkout was resumed. The carbon source was heated up to 1940 K, approximately 60 K below the target temperature. Excessive input power was required. Additional steps were taken to attain target temperature and higher efficiency from the filament: additional thermal radiation shielding was installed; the flux concentrator was reinforced and increased in size; and, a more tightly wound filament was installed.

In the meantime, the turbomolecular pump, which earlier broke down, was repaired and reinstalled. Figure 5, the carbon source subassembly, shows the carbon disc supported directly above the reworked filament. Figure 6 shows the emitter button/heat choke subassembly and the carbon source subassembly mounted together.

The emitter button/heat choke subassembly, the carbon source subassembly and the guarded electrode subassembly were installed and pumped down in the vacuum chamber. A schematic of this test setup is shown in Figure 7. The

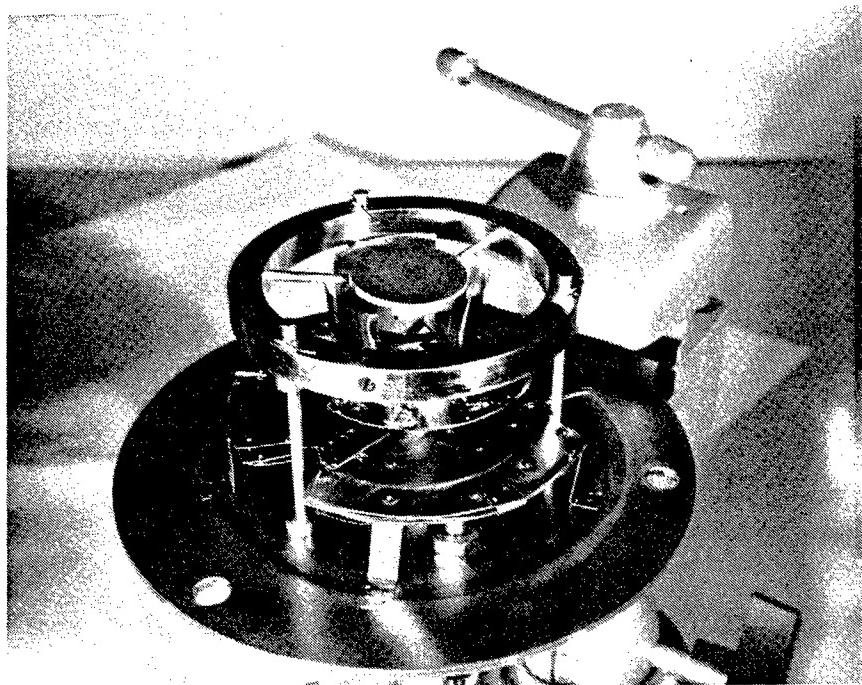


Figure 5. Carbon source subassembly.

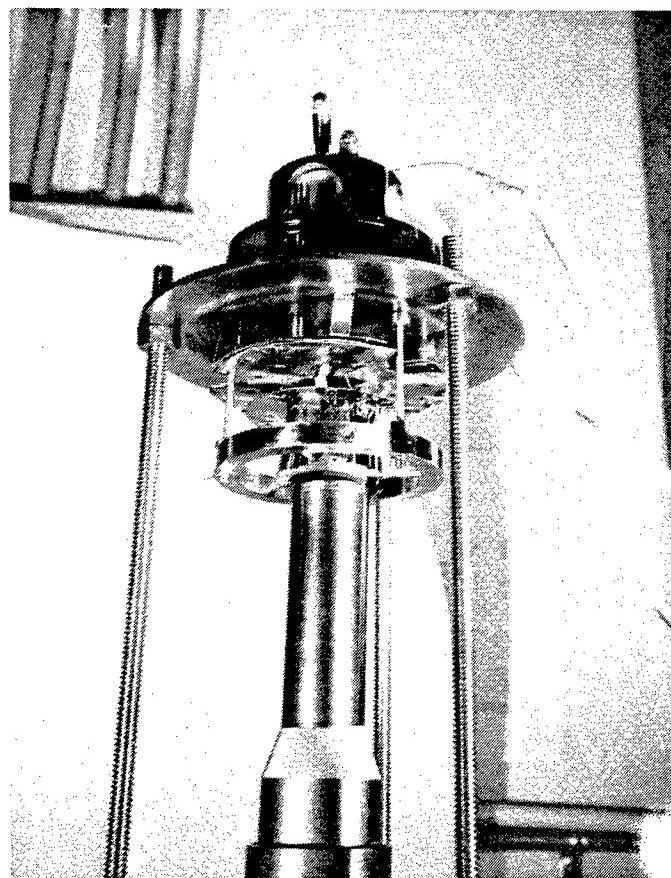


Figure 6. Carbon evaporation test assembly.

carbon source subassembly was heated up by the reworked filament. The emitter was able to reach 1750 K at a reasonable input power. Upon cooldown and inspection, it was discovered that a braze in the emitter heat choke had melted. Apparently, the emissivity of the heat choke was less than that used in thermal calculations resulting in less actual temperature drop in the heat choke than predicted. This caused a high temperature in the heat choke which led to melting the braze in question. Additionally, tack welds which attached the guard ring to the sheath insulator, were found to be cracked. The higher temperature heat choke heated the guard ring beyond its design temperature. The temperature difference between the guard ring and the sheath insulator fractured the tack weld.

In order to use most of the existing test article, a different approach to the test was conceived. The original concept envisioned heating the carbon source, which in turn heated the emitter button. A guarded electrode subassembly was located within the emitter/heat choke subassembly and would be able to make real time work function measurements of the back side of the emitter button.

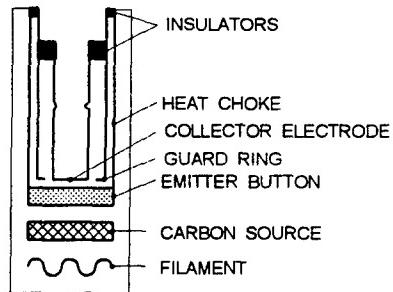


Figure 7. Test schematic.

A revised experimental concept is shown in Figure 8. The majority of time in the experiment is spent heating the carbon source, allowing carbon to evaporate onto the emitter and then diffuse through the emitter button thickness. During this phase, the experiment is set up as shown in Figure 8A. The carbon source is

heated by the filament which in turn heats the emitter button. A shortened emitter heat choke would be positioned such as to become a radiation shield for the carbon source, limiting radial thermal losses.

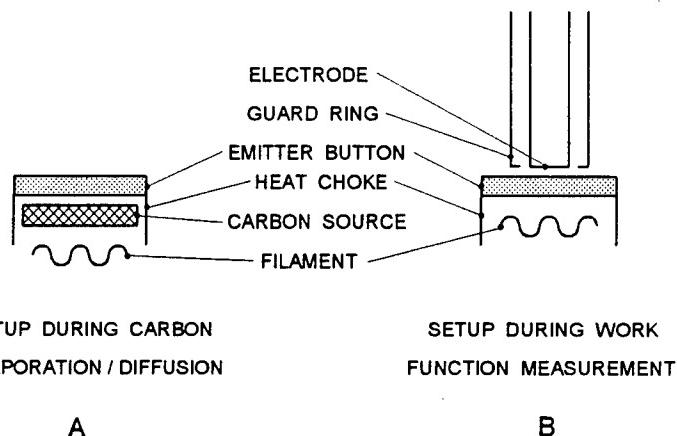


Figure 8. Revised experimental setup.

In order to perform periodic work function measurements, the experimental setup is changed to that shown in Figure 8B. During work function measurements, the carbon source would be removed and the emitter would be heated directly by the filament. The guarded electrode subassembly will be placed over the emitter/heat choke subassembly, permitting work function measurements to be made. In order to get measurable emission, the emitter would be heated above the temperature that it normally operated.

The revised filament, carbon source and emitter button/heat choke were reassembled and placed in the vacuum work station. The carbon source was heated to 2290 K and the emitter button achieved 1880 K with moderate heater input power. During this period, the Russian single-crystal tungsten disc was received and inspected during June. An EDAX analysis revealed that the disc was indeed pure tungsten. An x-ray diffraction analysis revealed that the crystal orientation was [110], as specified.

4.0 TEST RESULTS

After the experimental setup was checked out in the configuration of Figure 8A, the experiment was changed to the configuration shown in Figure 8B and a work function measurement was made. The measured work function for the polycrystalline tungsten disc was 4.27eV, with a standard deviation of $\pm 0.0619\text{eV}$, at $T \leq 1900\text{ K}$.

After work function measurement, the system was reconfigured to the configuration shown in Figure 8A. The assembly was heated up, with the carbon source operating at 2143 K and the emitter reaching equilibrium at 1843 K.

Operation continued for 188 hrs. At that time, the small rods supporting the carbon button had sagged such as to allow the carbon source to short out against the filament. These were repaired and heating was resumed. The carbon source was operated at 2131 K with the emitter reaching equilibrium at 1844 K.

At the end of 430 hrs the test setup was reconfigured to that shown in Figure 8B and work function measurements were made. A work value of $4.31 \pm .03\text{ eV}$ was measured. This compares to a value of $4.27 \pm .06\text{ eV}$ which was measured at the beginning of the test. Taking in account the measurement error band, virtually no change in the emitter work function had been observed at that time.

5.0 DISCUSSION AND CONCLUSIONS

Table 1 compares the STAR-C operating conditions to the test conditions experienced.

Because of the higher carbon temperature in the test, the carbon flux into the tungsten is higher in the test than that experienced during actual STAR-C conditions. The carbon diffusion rate is so high in polycrystalline tungsten at these temperatures that whatever carbon arrived on the surface would quickly diffuse across the tungsten thickness. The carbon concentration would be fairly uniform throughout the tungsten matrix and virtually no carbon gradient would be established.

Table 1. Component temperatures: experiment vs STAR-C conditions.

	STAR-C	EXPERIMENT
CARBON	2000 K	2131 K
EMITTER	1863 K	1844 K

Figure 9 shows the predicted carbon concentration in tungsten vs time, for two conditions. The steep line represents the carbon concentration at the accelerated operating conditions of this test. After 430 hrs (17.9 days) of operation, the predicted carbon concentration in the tungsten test button is 0.0075 g/cm^3 . Under the STAR-C operating conditions, with the surface of the core operating at 2000K, it would be expected to take 6120 hrs (255 days) of operation to achieve this same carbon concentration.

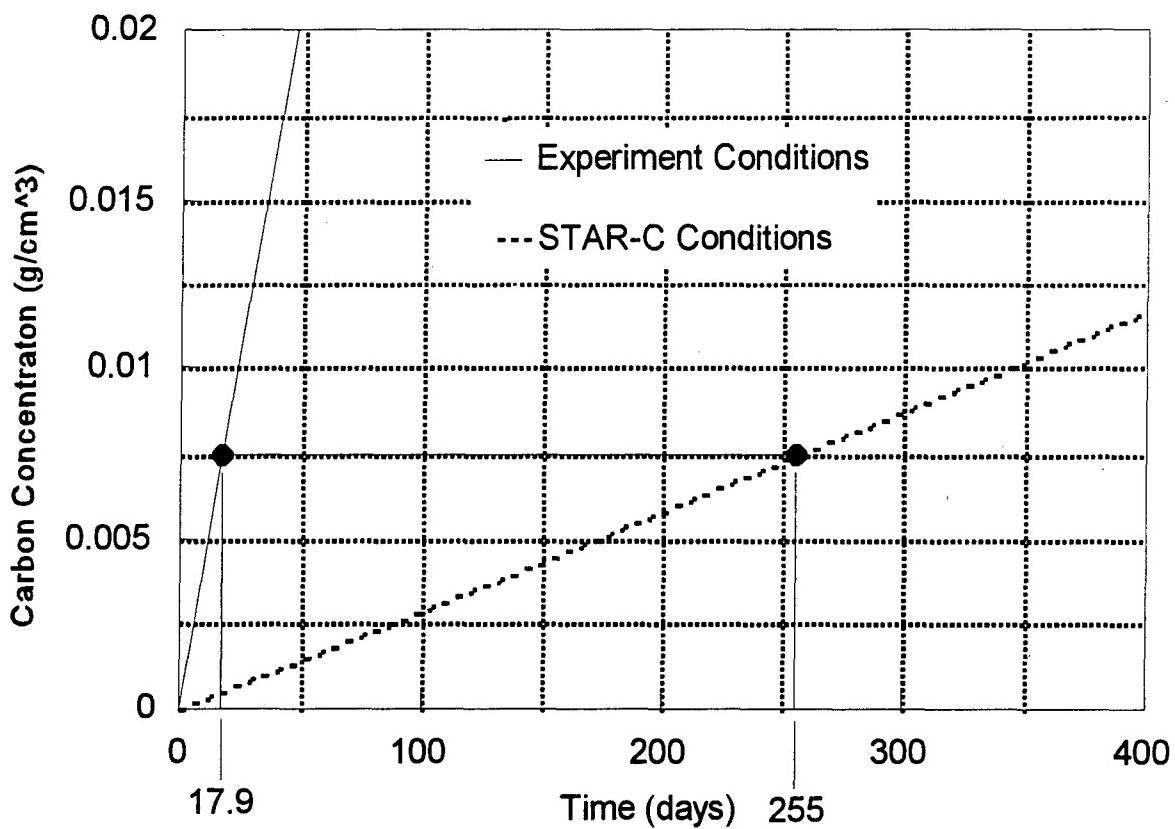


Figure 9. Carbon concentration vs time:
experiment vs STAR-C conditions.

No detectable change in work function was measured after 430 hrs at accelerated conditions. This indicates that the STAR-C system, using a conventional CVD polycrystalline emitter, could be expected to operate for 255 days without any noticeable degradation in performance due to carbon diffusion.

DISTRIBUTION LIST

AUL/LSE Bldg 1405 - 600 Chennault Circle Maxwell AFB, AL 36112-6424	1 cy
DTIC/OCP 8725 John J. Kingman Rd, Suite 0944 Ft Belvoir, VA 22060-6218	2 cys
AFSAA/SAI 1580 Air Force Pentagon Washington, DC 20330-1580	1 cy
PL/SUL Kirtland AFB, NM 87117-5776	2 cys
PL/HO Kirtland AFB, NM 87117-5776	1 cy
Official Record Copy PL/VTVP/Dr. Michael Schuller Kirtland AFB, NM 87117-5776	2 cys
PL/VT Dr Hogge Kirtland AFB, NM 87117-5776	1 cy